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UNITED STATES DEPARTMENT OF AGRICULTURE
BULLETIN No. 283
Contribution from the Bureau of Soils
MILTON WHITNEY, Chief

Washington, D. C. PROFESSIONAL PAPER September 28, 1915

THE PRODUCTION OF SULPHURIC ACID AND A PROPOSED NEW METHOD OF MANUFACTURE

By

WILLIAM H. WAGGAMAN, Scientist in Fertilizer Investigations

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THE PRODUCTION OF SULPHURIC ACID AND A PROPOSED NEW METHOD OF MANUFACTURE.

By WILLIAM H. WAGGAMAN, *Scientist in Fertilizer Investigations.*

INTRODUCTION.

The importance of sulphuric acid in science, arts, and manufacture has been increasing steadily for many years. Although scarcely any industry exists which does not employ this acid either directly or indirectly in the manufacture of its product,[1] the bulk of the sulphuric acid produced, both in this country and abroad, is used in the manufacture of fertilizer materials.

Since Liebig first proposed the treatment of bones or phosphate rock with sulphuric acid in order to render the phosphoric acid present water soluble, superphosphate has been the basis of the fertilizer industry, and the economic production of sulphuric acid has been the aim of numerous investigators and chemical engineers. The production of sulphuric acid of various strengths in the United States for the past three years, according to the figures of the United States Geological Survey, is given in Table I.

Because of the difficulty in shipping such a commodity all of the sulphuric acid produced is consumed in this country. Some of the products manufactured therefrom are shipped abroad but the quantity of acid entering into them is but a small percentage of the total production.

In 1913 the United States consumed 1,931,468 short tons of phosphate rock. Since practically all of this was made into acid phos-

[1] Phalen, W. C. Mineral Resources (1913).

phate, at least 2,000,000 tons or over 56 per cent of the total sulphuric acid produced was thus consumed. A large tonnage of acid is also consumed annually in the manufacture of ammonium sulphate, a by-product of the coking and gas industries, and also in the production of what are known to the fertilizer trade as "base goods," consisting of acidulated hair, wool, tankage, or other nitrogenous refuse of the packing industry.

TABLE I.—*Production of sulphuric acid in the United States for the years 1911, 1912, 1913, by grades.*

Year and grades.	Quantity.	Value.	Price per ton.
1911:	*Tons.*	*Dollars.*	*Dollars.*
50° Baumé	1,026,896	5,447,958	5.31
60° Baumé	421,165	2,624,042	6.23
66° Baumé	751,541	9,176,297	12.21
Other grades	10,728	121,575	11.33
Total and average	2,210,330	17,369,872	7.86
Total reduced to 50° Baumé	[1] 2,688,456	[1] 17,313,822	6.44
1912:			
50° Baumé	1,047,483	5,378,411	5.13
60° Baumé	451,172	2,727,764	6.05
66° Baumé	774,772	9,360,630	12.08
Other grades	66,166	871,214	13.17
Total and average	2,339,593	18,338,019	7.84
Total reduced to 50° Baumé	[2] 2,876,000	[2] 17,572,837	6.11
1913:			
50° Baumé	1,643,318	9,212,917	5.61
60° Baumé	509,929	3,202,528	6.28
66° Baumé	797,104	9,282,422	11.65
Other grades	63,158	986,659	15.62
Total and average	3,013,509	22,684,526	7.53
Total reduced to 50° Baumé	[3] 3,538,980	[3] 22,366,482	6.32

[1] Exclusive of acids of strength greater than 66° Baumé.
[2] Exclusive of electrolyte and acids of strength greater than 66° Baumé.
[3] Exclusive of 22,947 short tons of fuming acid, not convertible, valued at $318,044.

METHODS OF MANUFACTURE.

There are two general methods employed in the manufacture of sulphuric acid, namely the "contact process" and the "lead-chamber method." It is not within the scope of this paper to discuss in detail the numerous modifications of these two methods, but classified lists of the patents on the subject, together with short abstracts thereof, are given at the end of this paper.

THE CONTACT PROCESS.

Briefly the contact process consists in passing a purified mixture of air and sulphur dioxide derived either from sulphur or burning pyrites over some catalytic agent heated to dull redness, thereby effecting the further oxidation of the sulphur dioxide. The resulting sulphur trioxide is then usually absorbed in sulphuric acid, producing a very concentrated product.

Theoretically this should prove the simplest, cheapest, and most efficient method of making sulphuric acid, but in actual practice there are several details encountered both in the construction and running of a plant which unless given careful consideration will seriously affect the yield of acid and the cost of production.

Many different catalytic agents have been tried (notably platinum, palladium, iridium, the oxides or sulphates of iron, copper, chromium manganese, and silver, as well as the oxides of some of the rarer elements) in effecting the oxidation of sulphur dioxide, but no matter what catalyzer is used its efficiency is seriously impaired and often destroyed unless very elaborate systems are employed for purifying the gases before admitting them into the oxidation chamber. For instance, finely divided platinum (platinum black) has proved the most efficient catalyzer so far discovered, but the catalytic power of this body in bringing about the union of sulphur dioxide and air is seriously affected by the smallest traces of arsenic in the gaseous mixture. When pyrites are used as a source of sulphur dioxide, the arsenic which this mineral nearly always contains, passes over with the furnace gases and it is only by repeated washing and filtering that the last traces of this element can be removed.

Contrary to general opinion, therefore, a contact plant is both elaborate and costly and strict supervision by a competent chemical engineer is necessary to insure the best results.

The contact process has distinct advantages over the lead-chamber method where a pure concentrated acid is required, but in the manufacture of ordinary sulphuric acid (50° to 60° B.) for the fertilizer industry or for other purposes where the purity of the product is not essential, the latter method still holds first place (at least in this country) for efficiency and low cost of production.

THE LEAD-CHAMBER PROCESS.

The lead-chamber process with its various modifications has been fully treated by Lunge.[1] This author, as well as numerous other investigators, notably Weber, Winkler, Rascheg, Meyer, Pratt, Gilchrist, Falding, and Wedge, has described details of construction, methods of accelerating the chamber reactions, and proposed and discussed schemes for increasing the efficiency and lowering the cost of acid systems. It is thought, however, that a brief general description of the chamber process will help toward a better understanding of the modification of the process proposed in this paper.

In this country nearly all of the sulphuric acid is made from pyrites. The lump ore is imported chiefly from Spain, while the "fines" are a domestic product mined in Virginia, Georgia, Tennessee, and California. If the lump ore is used it is burned in brick

[1] Treatise on the Manufacture of Sulphuric Acid and Alkali, vol. 1.

furnaces having grates composed of single square bars which can be turned on their longitudinal axes to let the cinders down into the ash pits. Such furnaces hold from 3 to 5 tons each and are arranged in batteries of 20 to 25 for each set of lead chambers. The daily charge for each furnace when the system is in operation is from 750 to 1,000 pounds of pyrites. The burners for the pyrites "fines" consist of cylindrical furnaces having a series of shelves so arranged that the burning material can be mechanically raked from shelf to shelf until the fully burned cinder is discharged at the bottom of the furnace. The rakes are attached to a central air or water cooled shaft. In one type of furnace the shaft revolves; in another the shaft is rigid while the furnace itself revolves.

The gases from the pyrites burners are forced into a dust chamber fitted with baffle plates where the oxides of iron, arsenic, lead, zinc, etc., are in a large measure removed. From the dust chamber the gases enter the Glover tower, which consists of a lead tower (usually from 20 to 30 feet high and 6 to 8 feet across) lined with acid-resisting brick and partly filled with quartz or other acid-proof material so arranged that the dilute nitrous vitriol which is distributed from an apparatus at the top of the tower will trickle down through the interstices. The heat of the burner gases which enter the Glover tower at a temperature of from 300° to 400° C. drives off water and the oxides of nitrogen from the nitrous vitriol, restoring them to the system. The uses of the Glover tower therefore are threefold: first, to cool the furnace gases before allowing them to enter the lead chambers; second, to restore water and the oxides of nitrogen to the system; and third, to produce an acid more concentrated than that formed in the lead chambers.

From the Glover tower the gases enter the first of the lead chambers where most of the sulphuric acid is made. The lead chambers usually consist of large square or oblong boxes [1] made of sheet lead (weighing from 6 to 8 pounds per square foot) and having a capacity of from 25,000 to 75,000 cubic feet. Water in the form of fine spray or steam is introduced into the chambers at various points. This decomposes the nitrosulphuric acid formed into sulphuric acid and returns the oxides of nitrogen to the system to be again acted upon by the furnace gases. The number and size of the chambers used vary from 2 to 10 or more, depending on the number and size of the pyrites burners. Where the quantity of sulphur burned daily is large the acid plant is often divided into separate units, each battery of burners furnishing gases to its own set of lead chambers. The gases pass from the first to the second chamber and so on through

[1] In the Meyer Tangent system the lead chambers are cylindrical in form, while in the Falding system their height is several times their length and width.

the system, sulphuric acid being formed till the sulphur dioxide is practically exhausted.

The residual gases, consisting of nitrogen, oxides of nitrogen, some oxygen, and a small percentage of sulphur dioxide, then enter the lower part of the Gay-Lussac or recovery tower, which is similar in construction to the Glover tower, except that it is usually taller and wider (from 40 to 50 feet high and 8 to 15 feet across) and filled with coke instead of quartz. Strong sulphuric acid (1.5 to 1.7 specific gravity) trickles down the tower, absorbing the oxides of nitrogen from the residual gases which ascend through the coke column, and are finally discharged through a stack. The nitrous vitriol formed is then pumped to the Glover tower, diluted with water, and distributed as previously described.

MEASUREMENT OF A PLANT'S EFFICIENCY.

The efficiency of a lead-chamber plant is measured, first, by the amount of chamber space required for each pound of sulphur burned in 24 hours and the amount of acid (50° or 60° B.) made therefrom, and, second, by the amount of niter consumed or lost in the production of this acid.

Practically all sulphuric acid authorities agree that, provided the gases are present in the proper proportions, the two most important conditions necessary for efficient production are a thorough mixing of the gases and the control of their temperature.

The importance of the first of these conditions is self-evident, since in order to bring about complete chemical reaction the reacting substances must be in intimate contact with one another. The second condition is important because too low a temperature lessens the chemical activity of the gases, while a temperature above 100° C. prevents the condensation of water which it is claimed is necessary to bring about the decomposition of nitrosulphuric acid, an intermediate compound formed from the oxides of nitrogen in the system.

Numerous schemes to control these conditions have been devised, some of which have features of considerable interest and practical importance. While it is impracticable in a paper of this length to discuss in detail all of these processes, several that have been tried, apparently with some success, are described below.

METHODS FOR ACCELERATING THE CHAMBER REACTIONS.

Walter and Boeing [1] advocate the use of several hollow acid-proof partitions built across the chambers and so arranged that the gases enter the compartments through large holes near the bottom and are discharged from holes near the top. Numerous other small holes

[1] German patent No. 71908.

allow the admission and exit of the gases, thereby causing them to mix intimately without seriously interfering with the draft.

Because of the doubtful stability of these inner walls and the serious damage caused on their collapse this method is no longer used.

Gossage [1] as well as several other investigators proposed filling the chambers with coke so that the gases would be obliged to work their way through the interstices and thereby become thoroughly mixed. This scheme, however, has been abandoned because of the impurities introduced into the acid by the coke and the tendency of the coke columns to press against the lead walls, causing them to bulge and even break. The lack of any cooling device in this process also caused excessively high temperatures in the chambers.

Verstrart's [2] plan is similar to the above, except that stacks of bottomless stoneware jars filled with coke are used. The oxides of nitrogen are supplied to the system by allowing nitric acid to trickle down one of the stacks.

In Pratt's [3] process, which is much used in the Southern States, the gases are drawn through the first chamber by means of a fan, then through a tower packed with quartz down which flows dilute sulphuric acid, and finally they are reinjected into the front of the first chamber by means of the same fan. This circulatory system seems quite efficient and a number of plants where the process is employed are operating on less than 9 cubic feet of chamber space per pound of sulphur burned in 24 hours.

Meyer's [4] tangental chambers are designed both to mix and to cool the reacting gases at the same time.

The chambers are cylindrical in form, the first having water-cooled lead pipes suspended around the circumference. The gases are admitted at a tangent near the upper part of the chamber walls and are discharged from outlets in the centers of the chambers' bottoms. The gases are thus given a spiral motion which tends to mix them thoroughly while the water-cooled lead pipes reduce their temperature.

There are three installations of this type of plant in the United States. One at least is reported to have given great satisfaction.

Hartmann [5] obtained an increased yield of acid in the lead-chamber process by placing vertical, air-cooled lead pipes in the chambers. The chamber bottom is turned up around the lower ends of these pipes, forming hydraulic seals, and thus obviating the necessity of joints in the bottom of the chamber.

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[1] Lunge, Treatise on Manufacture of Sulphuric Acid, 1 Pt. I, p. 475.
[2] Bull. Soc. encour. ind. nat., 1865, p. 531.
[3] U. S. patents Nos. 546, 596, 652, 687.
[4] English patent No. 18376: Zeit. für ang. Chem. (1900), p. 742.
[5] Chem. Zeit., 1897, p. 877.

Blau [1] proposed to cool the gases in the first chamber by injecting a spray of cold sulphuric acid, and in order to obtain the optimum yield of acid from the gases in the subsequent chamber their temperature is raised by injecting sprays of warm sulphuric acid.

Falding's process [2] has for its object the segregating of the active gases in a system. To accomplish this he employs a chamber the height of which is approximately one and one-half times greater than its horizontal dimensions. The burned gases after passing through the Glover tower in the usual way are introduced either near the top or lower down on the chamber's side. Since the fresh gases are hot, not only because they have recently issued from the pyrites burners but because of the reactions taking place between some of the constituents, they collect in the upper part of the chamber in a relatively active layer.

As the reactions subside the spent gases gradually cool and settle to the bottom of the chamber, where they are withdrawn. Falding claims that by using the high chamber a zone of great chemical activity is always maintained in the upper part of the chamber, and that the spent or inactive gases, which in ordinary chamber systems act as diluents, are continually being removed from the active zone. It is also claimed that much less chamber space is required to complete the reactions by this process, so that even where large volumes of gases are handled each chamber is a unit in itself, being connected directly with the Glover tower instead of in series as in ordinary chamber systems.

A number of plants in this country are equipped with chambers of this type and it is reported that the process is commercially successful.

The main objections to the Falding system, in the opinion of the writer, are, first, that no provision is made for obtaining an intimate mixture of the gases other than the preliminary mixing brought about in the Glover tower, and, second, that no adequate means is provided for the condensation of the acid mist formed by the reactions.

The most widely used method of mixing and cooling the reacting gases is by means of intermediate towers containing plates, tubes, or baffles of some acid-resisting material cooled either by water, air, or dilute sulphuric acid. A number of different types of towers have been designed, but mention is here made of only a few of the better-known designs.

Lunge's plate tower [3] consists of a shell of lead either cylindrical or angular in form and filled with a series of perforated plates laid

[1] German patent No. 95083.
[2] U. S. patent 932771 (1909).
[3] Treatise on Sulphuric Acid, vol. 1, Pt. I, pp. 478–498.

horizontally. Each layer of plates is supported at some distance above the other by bearers in such a way that every plate is independent of the others. The plates are so constructed and placed that the holes in those of one layer do not come directly above the holes in the next layer below.

Dilute sulphuric acid is allowed to trickle down the tower, splashing from one layer of plates to another and meeting the hot-chamber gases as they wind upward through the tower. The film of dilute acid over the plates presents an immense cooling surface to the gases and at the same time furnishes the water necessary for the decomposition of the nitrosulphuric acid. The formation of this latter compound is, according to Lunge, a necessary link in the chamber process.

Gilchrist's pipe-column system[1] consists of lead towers (3 or 4 feet across and 15 feet high) having corrugated lead tubes, open at both ends, running through them horizontally like a steam boiler. The sides of the towers are boxed in with boards so as to form an air shaft which terminates in a flue at the top.

The chamber gases, together with water vapor, enter the pipe columns at the sides near the bottom and work their way upward through the towers. Contact with the air-cooled corrugated tubes condenses the sulphuric acid, which then drips in showers from one series of pipes to another and frees the oxides of nitrogen, restoring them to the system. The gases issue from the top of the towers and enter the next chamber, from which they are drawn into another series of pipe columns, and so on through the system till their oxidation is practically complete.

While some of the methods just described are designed to cut down the amount of chamber space required, none of them, with the exception of Falding's process, reduces the initial cost of erecting an acid plant; for, while less lead may be employed in constructing the chambers, the expense of the cooling and mixing towers more than offsets the saving in chamber material.

Another objection to most of the accelerating devices discussed above is that in order to mix the gases thoroughly they must be drawn or forced through small openings or made to pursue a meandering course by means of baffles or some acid-proof packing material in the towers. Under such conditions dust or impurities may clog the apparatus, choking off the draft and making it necessary to clean out the tower or chamber before operations can be resumed. Moreover, the collapse or disarrangement of the packing material within the tower may cause even more serious trouble.

Nearly all modifications of the chamber process complicate somewhat the running of an acid plant, and should therefore be constantly under the supervision of a competent chemical engineer.

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[1] Jour. Soc. Chem. Ind., 18, 459-462 (1899).

In the new modification of the chamber method described in this publication a complete mixture of the gases and the control of their temperature is brought about without the use of expensive or complicated apparatus and with practically no danger of clogging the system. While this method has been tried out only in the laboratory, and some of the analytical data are not altogether satisfactory, the results obtained prove that the principle is good and that the process, if worked on a factory scale, would probably be commercially successful.

In a review of the patent literature on the subject an apparatus was found which is somewhat similar to the one herein described. This United States patent (No. 446060) was taken out by E. and J. Delplace in 1891 and consists of a lead chamber having the shape of a ring with a sector cut out. The chamber is provided with two gas inlets at unequal distances from the center and contains at intervals distributing pipes leading from the upper part to the lower part of the chamber, so that the hot gases can be more thoroughly mixed with the cooler. The inventors state that in such a chamber the constant change in the direction of the gases and their impinging on the sides of the chamber cause a thorough mixture and a condensation of the acid formed. In this patent the right is reserved to vary the shape of the chamber provided the gases are led through a circular route.

The main objections to the above apparatus, in the opinion of the writer, are, first, that the chamber as described is of such a size that there must be spaces therein where the gases are relatively inactive; second, the pipes for conveying the hotter gases from the upper to the lower part of the chamber would hardly accomplish this unless they were the only route provided for the passage of the gases, and this according to the specifications is not the case; third, the use of pipes within the lead chamber unless they are cooled is always objectionable because of their excessive corrosion and the serious consequences resulting therefrom; fourth, a chamber of the shape described occupies an enormous amount of ground space. Where land values are high, this entails a large outlay for a factory site, as well as an expensive building to house the chamber.

NEW MODIFICATION OF THE CHAMBER PROCESS.[1]

This method is based on the fact that if a mixture of warm gases is drawn downward through a special flue their resistance to the downward pull, together with the constant change of their course, will tend to mix them very intimately, and unless the internal diameter of the flue is too great there will be practically no zones of inac-

[1] Work carried on under the direction of Dr. F. K. Cameron, to whom the author is indebted for much valuable assistance.

tivity in the apparatus. Moreover, the constant impinging of the gases on the walls of the spiral flue, which can be cooled either by air or water, makes it practicable to maintain the gases at a temperature most favorable for the efficient yield of sulphuric acid.

In the following laboratory experiments, however, the sulphur dioxide (SO_2) used was not directly derived from burning pyrites or sulphur, so it was necessary to heat the system artificially to attain a temperature as high as that obtained under factory conditions.

The sulphur dioxide was obtained from a small cylinder of the liquefied gas, which was weighed both before and after each experiment, and the SO_2 used thus determined. The oxides of nitrogen (chiefly N_2O_3 and N_2O_4) were produced by the action of dilute nitric acid on copper, and the rate at which they were used was roughly

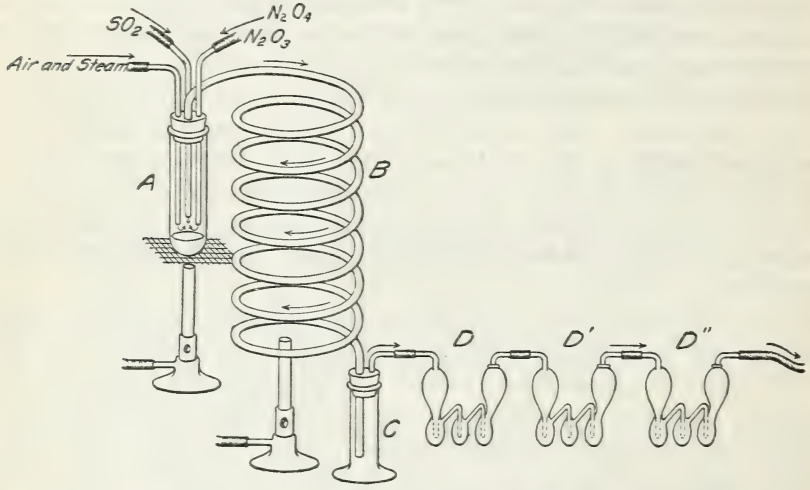

FIG. 1.—Apparatus used in proposed new method for manufacture of sulphuric acid.

determined by allowing the gases to bubble through dilute sulphuric acid saturated with these gases. A mixture of air and water vapor was obtained by drawing air through a flask of water heated to the boiling point.

The apparatus employed (fig. 1) consisted, first, of a large test tube (A), having a capacity of 200 c.c., and containing a little water heated to boiling. The oxides of nitrogen, sulphur dioxide, air, and water vapor were led to the bottom of this vessel by separate tubes and given a preliminary mixing. From the test tube the gases were drawn into the lead or glass spiral (B), which was heated to about 90° C. in order to facilitate the reactions. In winding downward through this spiral the warm gases were thoroughly mixed, with the result that most of the sulphuric acid produced in the system was formed in this coil. The residual gases were then passed through

the absorption bulbs (D, D′, D″) containing strong nitric acid, which absorbed the sulphur dioxide escaping oxidation in the spiral.

The quantities of sulphur dioxide converted into sulphuric acid in the tube (A) and the spiral (B), as well as that which escaped oxidation and was subsequently absorbed in the bulbs (D, D′, D″), were determined by analyses. The results of these analyses are given in Tables II and III.

When the lead spiral was used (Table II), the amount of sulphuric acid formed therein had to be determined by difference because of the formation of lead sulphate, which could not be entirely removed from the tube by washing.

On account of the many joints and rubber connections necessary in the apparatus there was some loss of sulphur dioxide in the system. When the lead spiral was used, the amount of this loss could not be determined, so all the errors occurring in Table II are thrown into column 5, making the figures for the sulphur dioxide oxidized in the lead spiral larger than they actually should be.

TABLE II.—*Sulphur dioxide oxidized to sulphuric acid in apparatus shown in figure 1. Lead spiral used and steady stream of oxides of nitrogen furnished throughout experiments.*

Number of run.	Time of run.	Rate per hour of SO_2.	SO_2 oxidized in system.		SO_2 lost in system.[1]	SO_2 escaping from end of spiral.
			In vessel A.	In lead spiral B.		
	Hours.	*Grams.*	*Per cent.*	*Per cent.*		*Per cent.*
5	2	6.2017	32.07	67.92	.....	0.01
7	2	6.4887	26.61	73.39	.....	Trace.
8	2	7.6569	23.00	76.99	.....	.01

[1] Could not be determined. Included in the figures in column 5.

An inspection of Table II shows that by passing sulphur dioxide, air, and water vapor through a lead spiral in the presence of an adequate supply of the oxides of nitrogen, the formation of sulphuric acid is practically complete, even when the gases are run at quite a rapid rate.

In ordinary chamber plants it is considered very good practice if only 10 feet of chamber space is required for every pound of sulphur burned in 24 hours. Figuring the chamber space required in run No. 8, it is seen that for every pound of sulphur burned in 24 hours only 0.139 foot of chamber space was required.

While it is hardly fair to compare the results obtained in the laboratory with those obtained on a factory scale, still the efficiency of the apparatus can be more readily judged by expressing the results in the conventional way.

The well-known characteristic of some metals, as well as metallic oxides and salts, of acting as catalytic agents made it seem possible

that the lead, lead oxide, or lead sulphate had something to do with the very efficient yield of acid obtained when using the lead spiral. Accordingly, a glass coil of the same length and internal diameter was substituted in the experiments shown in Table III.

TABLE III.—*Sulphur dioxide oxidized to sulphuric acid in apparatus shown in figure 1. Glass spiral used and steady stream of oxides of nitrogen furnished.*

Number of run.	Time of run.	Rate per hour of SO_2.	SO_2 oxidized in system.		SO_2 lost in system.	SO_2 escaping from end of spiral.
			In vessel A.	In glass spiral B.		
	Hours.	*Grams.*	*Per cent.*	*Per cent.*		*Per cent.*
20	1½	3.8311	14.17	75.14	3.15	7.34
19	1½	4.6404	43.09	50.18	3.20	2.63
12	2	5.7888	20.18	71.66	1.26	6.28

Table III shows that the yield of acid was not so great with a glass as it was with a lead spiral. In no instance was the gas run through the apparatus so fast as in the experiments in Table II, yet even though the oxides of nitrogen were present in large quantities in the gaseous mixture there was a loss of sulphur dioxide from the end of the spiral.

In order to try out the catalytic action of the lead coil, several runs were made with both the glass and lead spirals, but using no oxides of nitrogen. The results of these experiments are shown in Tables IV and V.

TABLE IV.—*Sulphur dioxide oxidized to sulphuric acid in apparatus shown in figure 1. Lead spiral used, but no oxides of nitrogen employed.*

Number of run.	Time of run.	Rate per hour of SO_2.	SO_2 oxidized in system.		SO_2 lost in system.[1]	SO_2 escaping from end of spiral.
			In vessel A.	In spiral B.		
	Hours.	*Grams.*	*Per cent.*	*Per cent.*	*Per cent.*	*Per cent.*
21	1	3.2642	0.40	38.08	.....	61.52
16	1	3.8459	.41	40.78	.....	58.81

[1] Could not be determined. Included in figures in column 5.

TABLE V.—*Sulphur dioxide oxidized to sulphuric acid in apparatus shown in figure 1. Glass spiral used, but no oxides of nitrogen employed.*

Number of run.	Time of run.	Rate per hour of SO_2.	SO_2 oxidized in system.		SO_2 lost in system.	SO_2 escaping from end of spiral.
			In vessel A.	In spiral B.		
	Hours.	*Grams.*	*Per cent.*	*Per cent.*	*Per cent.*	*Per cent.*
18	10	2.6713	(1)	0.75	[2] 10.02	89.23
15	10	2.8393	0.40	0.68	[2] 12.36	86.56

[1] Included in next column.

[2] The large loss of sulphur dioxide in the system was due in part to the renewal of the sulphuric acid in the wash bottle through which the gas was allowed to bubble before entering the system. This acid was not entirely saturated with SO_2 before these experiments were undertaken.

In comparing the results obtained in Tables IV and V it is evident that the lead spiral had some influence in the oxidization of the sulphur dioxide to sulphuric acid. The figures in column 5, Table IV, are obviously too high since all the errors due to the loss of gas in the system are thrown into this column. But while the sulphur dioxide and air was in each instance run through the lead spiral at greater speed than through the glass coil, the quantity escaping oxidation was much less in the former than in the latter case.

In order to determine if the catalytic action observed in the lead coil was due to lead or lead sulphate, two experiments were conducted using the glass spiral but no oxides of nitrogen. The conditions in these experiments were approximately the same as in those recorded in Table V except that in the first run the interior of the glass coil was coated with precipitated lead sulphate and in the second a lead chain was introduced into the glass spiral. The results are shown in Table VI.

TABLE VI.—*Sulphur dioxide oxidized to sulphuric acid in glass spiral coated with lead sulphate and in the same spiral after the introduction of a lead chain.*

Number of run.	Time of run.	Coil used.	Rate per hour of SO_2.	SO_2 oxidized in system.		SO_2 lost in system.	SO_2 escaping from end of spiral.
				In vessel A.	In spiral B.		
	Hours.		*Grams.*	*Per cent.*	*Per cent.*	*Per cent.*	*Per cent.*
22	1.0	(1)	2.6594	0.25	9.80	(2)	89.95
23	1.0	(3)	1.7523	0.09	53.60	(2)	46.31
24	1.5	(1)	2.5299	0.05	17.85	(2)	82.10

[1] Glass coil coated with lead sulphate. [2] Not determined. [3] Glass coil containing lead chain.

Here again, as in Tables II and IV, the amount of sulphur dioxide lost in the system could not be determined, but the results shown in Table VI indicate that while lead sulphate has some influence on the oxidation of sulphur dioxide, lead or lead oxide is a much more energetic catalytic agent. The presence of the oxides of nitrogen in the system, however, is necessary for the complete oxidation of sulphur dioxide to sulphuric acid.

FACTORY CONSIDERATIONS.

In the construction of a sulphuric acid plant along the lines of the apparatus described in this paper, it is proposed to dispense with the lead chambers and intermediate towers only. The lead spiral is not intended to replace the Glover tower, which is so important in the preliminary mixing and cooling of the furnace gases and in restoring the oxides of nitrogen to the system, nor is it intended to do away with the Gay-Lussac tower, which is essential for the recovery of these same oxides of nitrogen from the residual gases.

The amount of lead required per cubic foot of chamber space is considerably greater for a long spiral tube as herein suggested than for a cylindrical chamber in which the height and diameter are more nearly equal, but the great reduction in the chamber space required to produce sulphuric acid in the spiral should make it possible to build a plant with considerably less lead than is required in an ordinary chamber system. Moreover, the facts that the new type of plant requires no other device to accelerate the reactions and occupies much less ground space than the present type of factory, and therefore would not need large buildings, should decrease the initial cost of construction.

The cooling of the lead spiral would be accomplished largely by the air, but if necessary in hot weather streams of water could be played upon its upper portion. The water thus warmed by the heat of the reaction of the upper part of the spiral would tend to raise the temperature of the lower portion of the spiral where the reactions are not so vigorous. The immense amount of cooling surface contained in such a spiral, together with the constant movement of the acting gases, should also prevent excessive corrosion of the lead walls.

While it is not fair, and hardly practicable, to predict how efficient a plant built along the lines of the apparatus just described would prove, all the indications are that such a scheme, worked on a factory scale, would be economically successful.

In the following tables the author has attempted to classify all the American patents on the manufacture of sulphuric acid, both by the contact and chamber processes. While these classifications are by no means drawn along sharp and distinct lines, still they should be of considerable assistance in enabling one to pick out the particular phase of acid manufacture which interests him most.

In tabulating and abstracting these patents it is probable that numerous important points have been omitted, but in many cases this was unavoidable because of the limited space available in tables of this character. It is thought, however, that the information given will enable those interested in the subject to judge fairly well whether or not any particular patent is of sufficient value to him to warrant further investigation.

NOTE.—Application has been made for a patent covering the process here described; if patent is allowed, it will be donated to the people of the United States.

APPENDIX.

TABLE VII.—*Apparatus and processes designed primarily for the efficient oxidation of SO_2 used in the manufacture of sulphuric acid by the "contact process."*

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
636,924	1899	Schroeder, Max	Salt impregnated with platinum.	Process (no illustrations)	A mixture of platinum chloride and some other soluble salt is dissolved in water, the solution evaporated to dryness, and the platinum precipitated through the mass by heating.	Production of an efficient contact mass.
664,630	1900	Frasch, H. A	Ferric oxide and iron compound, dried air, and SO_2.	Process (1 illustration. A converter).	A ferric-oxide producing substance is burned in a converter on a ferric-oxide base. Thoroughly dried air and SO_2 are passed over the material which is constantly being renewed from above.	Maintenance of fresh contact substance.
681,698	1901	Hasenbach, W	Ferric oxide, dried air, and SO_2.	Process (no illustration)	Dried air is passed through burning pyrites and then the mixture of gases is led over burned pyrites.	Utilization of heat of burner gases and employment of cinder as catalyzer.
686,021	1901	Blackmore, H. S	Oxide of iron, S, and SO_2	Not described. Process (no illustrations).	SO_2 impregnated with S vapor is passed over pyrites cinders. SO_3 absorbed in usual way.	Production of SO_3 and FeS.
686,022	1901	do	Oxide of iron (supplementary to above).	Process (no illustrations)	SO_2 is passed over oxide of iron, reducing the latter. The oxide of iron is then regenerated by passing air over it.	Continual regeneration of contact substance.
687,834	1901	DeHaen, C. J. E	Vanadic acid	do	Mixture of air and SO_2 is passed over asbestos impregnated with vanadic acid.	Production of an efficient contact body.
690,133	1901	Clemm & Hasenbach	Pyrites cinder and platinized asbestos, SO_2, and air.	Apparatus (2 illustrations) consisting of two contact chambers.	Mixture of gases is first passed over pyrites cinders, then dry filtered and passed over platinized asbestos.	Economy in filtering devices and less need of cooling devices.
700,512	1902	Krauss & Von Berneck	Pyrites cinders impregnated with ferrous sulphate, SO_2, air.	Process (2 illustrations. Inclined rotating cylinder).	Pyrites cinders impregnated with ferrous sulphate are fed into the upper end of a rotating cylinder, meeting a current of air and SO_2 from the furnaces.	In hottest or lowest part of cylinder catalysis occurs, in next or middle zone $FeSO_4$ is decomposed evolving SO_3, and in the third or coolest zone SO_2 is fixed and returned to the system.
711,186	1902	Stone, G. C	Separate compartments containing catalyzer, SO_2, air.	Apparatus (2 illustrations) consisting of a series of compartments containing the catalytic agent. Compartments can be renewed and replaced.	Gaseous mixture is passed through several chambers each of which contains some of the catalytic agent.	An efficient apparatus for renewing the contact body.

TABLE VII.—*Apparatus and processes designed primarily for the efficient oxidation of SO_2 used in the manufacture of sulphuric acid by the "contact process"*—Continued.

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
716,985	1902	Clemm, A.	Clay impregnated with copper sulphate, SO_2, air.	Process (no illustrations)	A mixture of SO_2 and air is led over unburnt clay impregnated with crystallized $CuSO_4$.	Efficient catalytic agent.
729,735	1903	Clemm & Hasenbach.	Oxides of copper and chromium, platinum black, SO_2, air.	do	Gaseous mixture is first passed over oxides of copper and chromium, then through some porous medium to filter out impurities, and finally over platinized asbestos.	Efficient method of making SO_3 and H_2SO_4.
752,165	1904	Hasenbach, W.	Platinized fabric, SO_2, air.	Apparatus (4 illustrations). Chamber fitted at intervals with sheets of platinized asbestos.	A mixture of air and SO_2 is passed through a number of sheets of platinized fabric.	More efficient oxidation of SO_2, due to intimate contact of the gases with the catalyzer.
758,844	1904	Lunge & Pollitt.	Pyrites cinders impregnated with arsenic, SO_2, air.	Process (no illustrations)	A mixture of air and SO_2 is passed up through pyrites cinder, the lower part of the column being rich in arsenic.	More efficient catalyzer.
769,585	1904	Blackmore, H. S.	Refrigeration, gold plated copper gauze, electric current, SO_2, air.	Apparatus (6 illustrations) consisting of chamber fitted with gold-plated wire gauzes, electrically heated.	A refrigerated mixture of air and SO_2 is passed up through a series of gold-plated copper gauzes heated by an electric current.	Apparatus which obviates the necessity of a catalytic agent and prevents dissociation of SO_3 formed.
774,083	1904	Knietsch, R.	Platinized asbestos on horizontal perforated plates, SO_2, air.	Apparatus (4 illustrations). A tube containing a series of perforated plates on which platinized asbestos is spread.	The gaseous mixture is passed up through the tube fitted at intervals with horizontal perforated plates, on which is spread platinized asbestos.	A more efficient method of exposing the gases to the catalyzer.
778,099	1904	Blackmore, H. S.	Refrigeration, CO_2, SO_2, heat.	Process (6 illustrations). Chamber fitted with gold-plated wire gauzes electrically heated.	A mixture of CO_2 and SO_2 is passed through a series of electrically heated gauzes. The SO_2 is oxidized as follows $SO_2+CO_2=SO_3+CO$. The endothermic reaction CO_2 to CO more than counteracts the heat of the reaction SO_2 to SO_3.	Control of temperature and obviation of the necessity of catalytic agent.
823,472	1906	Knietsch, R.	Platinized asbestos	Apparatus (4 illustrations) for conducting process described in patent No. 774,083.	Apparatus for patent No. 774,083	An apparatus for the efficient oxidation of SO_2 to SO_3.
869,094	1907	Kitsee, I.	Static electric discharges, SO_2, air.	Apparatus (1 illustration) consisting of a chamber in which the gases are led through electrical discharges.	The gases are led upward through a chamber and subjected to the action of electrical discharges.	Efficient method of oxidizing SO_2.

930,471	1909	Hallock, W	Radioactive or some other ionizing agent, SO_2, air, water.	Apparatus (1 illustration). Flask containing rod coated with radioactive material.	A mixture of air, SO_2, and water vapor is passed through the flask.	Oxidation of SO_2 to H_2SO_4.
988,646	1911	McFetridge, J	Finely divided oxide of iron, SO_2, air.	Apparatus (2 illustrations). Rotating furnace for burning pyrites and oxidizing part of SO_2, formed. A chamber plant then used to oxidize remainder of SO_2.	Finely divided pyrites are blown into a rotating furnace. The SO_2 formed is partially oxidized by the pyrites cinders. The dust is settled out of the gases and the SO_2 absorbed. The residual gases are then treated by the "chamber process."	Utilization of pyrites to oxidize (partially) the SO_2 formed.
1,007,5	911	Beckman, J. W	Alundum, particles, 40 pounds; copper oxide, 9 pounds; silica, 0.7 pound.	Process (no illustrations)	The ingredients are heated to a temperature slightly above the melting point of copper, and the latter then reduced in films over the alundum grains.	Production of an efficient porous catalytic agent.
1,018,402	1912	Albert, Kurt	A combination of ferric peroxide with an alkaline earth FeO_2. SrO, SO_2, air.	do	Sulphur dioxide and air are passed over the compound at a temperature of from 450° to 550° C.	Production of a contact body of great efficiency not so sensitive to moisture as pyrites cinders.
1,030,508	1912	Eschellmann & Harmuth.	A number of layers of platinized asbestos, SO_2, air.	Apparatus (3 illustrations). A contact chamber readily accessible for cleaning purposes containing numerous layers of platinized asbestos.	The mixture of SO_2 and air is passed first through a rather thick layer of platinized asbestos, then through a perforated plate, and finally through 25 layers of platinized asbestos.	An efficient contact chamber designed to mix and oxidize.
1,036,610	1912	Grosvenor, W. M	Catalyzer contained in several compartments of rotating cylinder, SO_2, air.	Apparatus (4 illustrations). A revolving cylinder containing the catalytic agent in several compartments.	The gaseous mixture is admitted into the apparatus in such a way that the cool gases come into contact with the hottest part of the catalyzer.	An efficient oxidation chamber.
1,102,670	1914	von Keler & Weinder	Silver vanadium compound	Process (no illustrations)	A compound of silver and vanadium.	Production of an efficient catalytic agent.
1,103,017	1914	Ellis, C	Tellurium or selenium compounds.	do	The substance is distributed through pumice or some other suitable porous medium.	Do.
1,103,522	1914	Lihme, I. P	Metallic oxide capable of absorbing hydrogen compounds of arsenic, SO_2, air.	Process (3 illustrations)	The gases are first passed over metallic iron to reduce the arsenic and then over pyrites cinders to absorb the arsenic hydride.	An efficient means of removing arsenic from burner gases.

TABLE VIII.—*Apparatus and processes designed to control the temperature of the gases used in the manufacture of sulphuric acid by the contact process.*

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
652,119	1900	Knietsch, R.	Contact body, heat and refrigeration, SO_2, air.	Process (11 illustrations).	The gaseous mixture is heated by the reactions taking place in the contact chamber. The catalyzer, in turn, is cooled by the gases entering the apparatus.	Control of the temperature of reacting gases and product.
677,692	1901	Pauli & Krauss.	SO_2, air, contact body, heat, and refrigeration.	Process (2 illustrations).	do.	Utilization of heat of reaction to warm reacting gases. Cooling of SO_3 formed.
688,020	1901	Knietsch, R.	SO_2, air, contact body, heating and cooling of gases.	Apparatus (11 illustrations) for process covered by patent No. 652,119.	The apparatus used is varied according to the richness of the gases treated, but the general scheme is to cool contact chamber by surrounding it with chamber or tubes containing a cooling medium.	Apparatus for control of reacting gases and product.
688,469	1901	do.	do.	Various forms of apparatus described in patent No. 652,119.	do.	Do.
688,470	1901	do.	do.	do.	do.	Do.
688,471	1901	do.	do.	do.	do.	Do.
688,472	1901	do.	do.	do.	do.	Do.
690,062	1901	do.	do.	do.	do.	Do.
692,018	1902	do.	do.	do.	do.	Do.
719,332	1903	Herreshoff, J. B. F.	SO_2, air, contact body, heating and cooling of gases.	Process (1 illustration).	The gaseous mixture is heated, then brought into contact with part of catalyzer, then cooled and passed over more of the catalyzer.	More efficient yield of SO_3 under these conditions.
719,333	1903	do.	do.	Apparatus to bring about process described in patent No. 719,332.	do.	A more efficient apparatus for oxidation of SO_2.
723,595	1903	Ferguson, W. C.	SO_2, air, series of contact chambers, air added at intervals.	Apparatus (1 illustration) consisting of plurality of contact chambers with air inlets in connecting pipes.	The gaseous mixture is passed through a series of contact chambers, air being added after it emerges from each chamber.	More efficient yield of SO_3, due to cooling of gases and thorough mixing of same.
723,596	1903	do.	do.	Process (1 illustration) for apparatus described in patent No. 723,595.	do.	An apparatus to bring about the more efficient oxidation of SO_2.
726,076	1903	LeBlanc & Krauss.	SO_2, air, contact body, heat.	Process (1 illustration).	A mixture of 7 per cent SO_2, 9 per cent O, is conducted over a catalyzer at 500° C. at speed of 120 liters per minute per kilo of contact body, then through other contact chambers at successively lower temperatures.	More efficient oxidation of SO_2 due to higher temperature of first contact chamber.

731,758	1903	Daub, C.....	SO_2, air, series of contact chambers, heating or cooling of the gases.	Apparatus (4 illustrations). A number of contact chambers in an upright column, each chamber connected by pipes.	The gaseous mixture is led from one chamber to another, the temperature being controlled by heating or cooling the connecting pipes.	Better control of temperature and better yield.
736,876	1903	Raynaud & Pierron...	SO_2, air, 3 catalytic bodies of different oxidizing power.	Process (2 illustrations).....	The gaseous mixture is led successively through 3 vessels heated to uniform temperature and having different catalytic power.	Efficient oxidation of SO_2.
742,502	1903	Schroeder, M.....	SO_2, air, cooling and subsequent heating of the gases.	Process (5 illustrations).....	The gases are cooled and purified and then their temperature is again raised by the heat of the fresh burner gases.	Purification of gases and economy in fuel.
751,941	1904	Raynaud & Pierron...	SO_2, air, 3 contact chambers heated at different temperatures.	Process (2 illustrations).....	The gaseous mixture is conducted successively through 3 contact chambers, the first heated to 300° C., the second to 500° C., and the third to 400° C.	More efficient oxidation of gaseous SO_2.
792,205	1905	Eschellmann & Harmuth.	SO_2, air, catalyzer contained in tubes, cooling of gases.	Apparatus (2 illustrations) consisting of gas-receiving chamber and lower catalytic chamber.	The gaseous mixture is led first into a chamber to cool, then into a set of tubes containing the catalyzer. The SO_3 formed is cooled by the incoming gases.	Method of controlling temperatures of gases and oxidizing same.
793,543	1905	Schroeder, Max.....	SO_2, air, cooling and thorough mixing of gases.	Apparatus (3 illustrations) comprising a plurality of contact chambers, adjacent contact bodies being separated by mixing spaces and passageways.	In passing through the catalytic body the gases are made to mix thoroughly. Thus the heat of the interior of the gaseous stream is transmitted to the exterior or cooler portion.	Uniform distribution of heat and thorough mixture of gases.
809,450	1906	Knietsch, R.....	SO_2, air, catalyzer, cooling of gases.	Process (1 illustration).....	The gaseous mixture is passed into an oxidizing chamber, the resulting SO_3 absorbed, then the residual gases are passed into a second, and if necessary, a third chamber.	Economy in platinum as a contact body.
828,268	1906	Blackmore, H. S.....	Electric current, SO_2, NO_2, CO_2.	Process (6 illustrations).....	A mixture of SO_2, NO_2, and CO_2 is passed up through a column, the CO_2 being reduced and SO_3 formed.	Control of temperature by the endothermic reaction CO_2 to CO.
857,389	1907	Ferguson, W. C.....	SO_2, air, contact material arranged on shelves, cooling of gases.	Apparatus (2 illustrations) consisting of contact chambers provided with heat retaining covering of unequal effectiveness.	The gaseous mixture is passed through a relatively narrow tube containing shelves on which the contact body is spread.	Apparatus presenting large cooling surface and causing intimate contact of gases with catalytic agent.
1,036,473	1912	Eschellmann & Harmuth.	SO_2, air, catalyzer in 2 chambers.	Process (2 illustrations).....	The gaseous mixture is first passed through a relatively thin layer of the catalyzer distributed over a considerable surface, then at a more rapid rate through a thicker layer of the catalyzer.	Efficient temperature control and distribution of catalyzer.
1,036,609	1912	Grosvenor, W. M.....	SO_2, air, heat, platinum contact body.	Process (6 illustrations).....	The gaseous mixture is conducted by a circuitous route into the center of the contact mass and returns by another circuitous route adjacent to to the first.	Control of temperature through mixture of gases.

TABLE VIII.—*Apparatus and processes designed to control the temperature of the gases used in the manufacture of sulphuric acid by the contact process*—Continued.

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
1,099,530	1914	Wolff, T.....	SO_2, air, catalyzer, heating and cooling actions.	Apparatus (1 illustration) for cooling catalytic body and heating mixture of SO_2 and air.	The incoming gaseous mixture is passed through a preheater, the temperature of which is raised by the SO_3 formed in the contact chamber. The temperature of the gaseous mixture can be controlled by allowing only a portion to flow through the preheater.	Efficient method of controlling temperature.

TABLE IX.—*Apparatus and processes designed for the purification of the gases used in the manufacture of sulphuric acid by the contact process.*

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
90,067	1869	Bigelow, A., and Baldwin, S.	SO_2, air, decrease in velocity of gaseous current.	Process (2 illustrations).....	The burner gases are led into a chamber where their velocity is decreased, then into a second and larger chamber.	Scheme for settling the suspended material.
140,034	1873	Goodfellow, J. T., and Sabbaton, F.	SO_2, air, impeding of the gaseous current. Constantly renewed purifying agent.	Apparatus (7 illustrations) designed to impede the gaseous current and to renew purifying agent.	The gases are made to pursue a meandering course over the purifying agent which is being constantly renewed.	Efficient gas purifier.
670,559	1901	Hasenbach, W.....	Dried air, SO_2.....	Apparatus (1 illustration) for drying air before admitting it to pyrites burners.	The air used for the combustion of the pyrites is thoroughly dried before admitting to the burners.	Exclusion of moisture from the contact chamber.
711,187	1902	Stone, G. C.....	SO_2, air, refrigeration, and filtration.	Process (2 illustrations).....	The gaseous mixture from the furnace is first cooled and then the arsenic deposited on wire gauzes covered with some filtering device.	Removal of arsenic from burner gases.
711,188	1902	.....do.....	.....do.....	Apparatus (2 illustrations) for conducting process described in patent No. 711,187.	.....do.....	Do.
734,849	1903	Gin, G.....	Water, sulphuric acid, refrigeration, heat, SO_2, air.	Process (2 illustrations).....	The gases are first cooled, the SO_3 present is then removed by H_2SO_4, and the SO_2 is absorbed in water and then driven off by the heat of the burner gases, mixed with air, and admitted to the contact chamber.	Efficient yield of SO_3 with minimum danger to contact body.
739,108	1903	Rabe, H.....	SO_2, air, cooling and filtration.	Process (1 illustration).....	The gaseous mixture is first cooled, then filtered through a heavy layer of asbestos. It is then conducted over shelves containing bisulphite, and then up through a tower fed with H_2SO_4 (strong). The gases are then admitted to the contact body.	Efficient method of purifying gases.
758,222	1904	Stone, G. C.....	Condensation and filtration of burner gases.	Apparatus (1 illustration) consisting of several chambers for filtering and condensing gases.	The gaseous mixture is drawn down through several compartments, the chambers being alternately filters and condensers.	Efficient gas purifier.
793,745	1905	Shields, J.....	SO_2, air, constantly renewed filtering material.	Process (1 illustration).....	The gases are led into a heap of the filtering material which is so arranged that the gases constantly come in contact with fresher material.	Efficient method of purifying burner gases.

TABLE IX.—*Apparatus and processes designed for the purification of the gases used in the manufacture of sulphuric acid by the contact process*—Con.

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
798,216	1905	Scharff, M., and Slama, F.	SO_2, air, iron oxide, copper oxide, or chromic oxide, manganese dioxide, or mixtures of these oxides.	Process (no illustrations)	The gases are led over one or more of the oxides heated to redness.	Removal of arsenic from the gases.
798,302	1905	do	SO_2, air, incombustible granular bodies.	do	The gaseous mixture is passed through infusible granular bodies consisting of clay, coke, furnace slag, phosphates, and silicates of the alkalies heated to 350° to 400° C.	Purification of the burner gases.
822,373	1906	Knietsch, R.	SO_2, air, steam, cooling, washing, and drying.	Process (3 illustrations)	The gases are first treated with steam, then cooled and washed, and finally dried by passing over coke impregnated with sulphuric acid.	Removal of dust arsenic and other impurities.
891,703	1908	Jonas, O.	Acetylene, tetrachloride, SO_2, air.	Process (no illustrations)	The burner gases are passed through towers fed with acetylene tetrachloride.	Removal of arsenic.
891,775	1908	do	Dichlorbenzene, SO_2, air	do	The gases are passed through towers fed with dichlorbenzene.	Do.
900,500	1908	Eschellmann, G.	Coke filters and alkaline earth hydroxide, SO_2, air.	Process (1 illustration)	The gases are first passed through a cooler, then through 2 coke filters, then up through a tower fed with milk of lime.	Removal of impurities and chlorine.
922,516	1909	Rockliff, W., and Booth, J.	SO_2, air, centrifugal force	Apparatus (6 illustrations) consisting of a longitudinal spiral deflector plate contained in a horizontal flue.	The gases in passing through the flue are submitted to the centrifugal action of the spiral deflector which throws the dust and impurities against the sides.	Method of precipitating suspended particles.
931,868	1909	Hegeler, H., and Heinz, N.	SO_2, air, refrigeration and absorption of SO_3 from gases by H_2SO_4.	Process (3 illustrations)	The gases from the furnace are mixed with cool gases of the same content of SO_2. They are then passed through a tower fed with sulphuric acid and finally dried.	Efficient method of purifying gases containing SO_2.
937,147	1909	Eschellmann & Harmuth.	SO_2, air, coke filter	Process (1 illustration)	The gases contaminated with oil from the gas pump are passed through a coke filter.	Removal of oil from burner gases.
937,148	1909	do	do	Apparatus (1 illustration) consisting of coke filter for removing oil from burner gases.	do	Do.
940,596	1909	Herreshoff, J. B. F.	SO_2, air, refrigeration, filtration, and desiccation.	Apparatus (1 illustration) consisting of a dust chamber and a series of combined coolers and scrubbers.	The gases are led first into a dust chamber, then through the series of combined scrubbers and coolers, and finally through a dryer.	Production of an efficient gas purifier.

989,801	1911	Reise, C. L.....	SO_2, air, refrigeration, desiccation, lime.	Process (1 illustration).....	The gases are first cooled, then dried, and finally passed over lime (CaO).	Efficient method of removing Cl, HCl, H_2S_2, and H_2SO_4.
1,016,476	1912	Cottrell, F. G.....	SO_2, air, electrical precipitation.	Process (2 illustrations).....	The gases are lead through a flue containing a number of pairs of screens. The first screen of each pair is fed with dilute H_2SO_4 and the second with fuming H_2SO_4. An acid mist is thus formed which is precipitated by electric charges.	Precipitation of dust and impurities out of gases.
1,028,880	1912	Howard, H.....	SO_2, air, refrigeration, and strong sulphuric acid.	Process (1 illustration).....	The gases are first dried by cooling and then by passing through strong H_2SO_4.	Means of removing moisture without excessive use of sulphuric acid.
1,113,437	1914	Herreshoff, J. B. F....	SO_2, air, cooling action.....	Process (5 illustrations).....	The gases, after being washed with sulphuric acid (40°B.), are cooled from 200° to 100° F., thus precipitating much of the moisture.	Economy in acid necessary to dry gases.

TABLE X.—*Apparatus and processes designed to absorb the SO_3 formed in the manufacture of sulphuric acid by the contact process.*

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
695,180	1902	Stone, G. C.	SO_3, cooling action and dilute H_2SO_4.	Apparatus (4 illustrations) consisting of cooled tubes down which dilute H_2SO_4 flows. Tubes contain dams to delay flow.	SO_3 from contact chamber is led up through the cooled tube down which dilute sulphuric acid flows.	Efficient absorptive apparatus for SO_3.
722,981	1903	Herreshoff, J. B. F.	Sulphuric acid of constant strength, SO_3.	Process (1 illustration)	SO_3 produced in contact chamber is passed into sulphuric acid (90 per cent) maintained at constant strength by addition of water.	More efficient method of absorbing SO_3.
737,625	1903	do	SO_3, sulphuric acid of constant strength.	do	After absorbing SO_3 from the gases issuing from the contact chamber part of the acid is removed and the remainder automatically diluted and restored for absorptive purposes.	Automatic regulation of absorptive medium.
737,626	1903	do	do	Apparatus (1 illustration) for method covered by patent No. 737,625.	do	Do.
789,634	1905	Schroeder, M.	SO_3, dilute H_2SO_4 percolating through sand and quartz or other acid-proof material.	Process (2 illustrations)	SO_3 from contact chamber is passed up through a tower containing acid-proof packing over which dilute H_2SO_4 flows.	Intimate contact with the H_2SO_4 absorbs the SO_3 from gases.
800,218	1905	Knietsch, R.	SO_3, H_2SO_4 containing at least 27 per cent free SO_3.	Process (no illustrations)	SO_3 from contact chamber is passed into H_2SO_4 containing at least 27 per cent free SO_3. This acid does not attack iron.	Method of obtaining H_2SO_4 free from iron.
816,918	1906	do	SO_3, H_2SO_4 containing between 97 and 99 per cent H_2SO_4.	Process (1 illustration)	SO_3 from contact chamber is passed into H_2SO_4 containing from 97 to 99 per cent H_2SO_4. This acid does not attack iron.	Production of H_2SO_4 free from iron.
866,843	1907	Cottrell, F. G.	SO_3, water in form of spray.	Process (1 illustration); apparatus (2 illustrations). Centrifugal appliance consisting of rotating shell having plurality of concentrically placed cylinders.	SO_3 from contact chamber is sprayed with water, liquid particles of H_2SO_4 being formed.	Absorption of SO_3 and precipitation of H_2SO_4.
866,844	1907	do	do	do	do	Do.
894,792	1908	Eschellmann & Harmuth.	SO_3, absorptive medium, cooling action.	Apparatus (2 illustrations) consisting of a plurality of absorption chambers down the walls of which an absorbing liquid flows.	SO_3 from contact chamber is led through a plurality of absorption chambers adapted to contain an absorbing liquid. The walls of the chambers are cooled.	Efficient absorption apparatus.

1,002,824	1911	Cox, I. J.	SO_3, concentrated H_2SO_4, cooling action.	Process (1 illustration)	SO_3 from contact chamber is passed up through a coke column down which strong H_2SO_4 (cooled) flows. The acid then goes to cooler and is again distributed through the tower.	Acid for efficient absorption of SO_3.
1,013,638	1912	Briggs & Merriam	SO_3, H_2SO_4 (97 to 99.5 per cent) cooled.	do	SO_3 from contact chamber and air in passing up through an absorption tower meet cooled H_2SO_4 flowing down the tower. The temperature of the tower is regulated by a film of water flowing down the exterior walls of the tower.	A process for the efficient absorption of SO_3.
1,082,301	1913	do	do	Apparatus (1 illustration) for carrying out process described in patent No. 1,013,638.	do	An apparatus for the efficient absorption of SO_3.

TABLE XI.—*Apparatus and processes for the more efficient mixing of the gases used in the manufacture of sulphuric acid by the contact process.*

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
384,841	1888	Hamsch & Schroeder	Air and SO_2 under pressure, catalyzer.	Process (2 illustrations)	A mixture of air and SO_2 is compressed and passed over heated platinum.	Compression tends to mix gases and produce more efficient yield.
677,670	1901	Krauss & Wach	Air and SO_2, admitted at various points in system, catalyzer.	Apparatus (1 illustration) consisting of contact chamber with gas ingress at two or more points.	Air and SO_2 are admitted at various points in the contact chamber.	Control of temperature and efficient mixture of gases.
688,020	1901	Knietsch, R	SO_2, air, mixing and cooling.	Apparatus (2 illustrations)	The gaseous mixture is passed through a number of small pipes.	Apparatus for obtaining an efficient mixture and controlling temperature of gases.
688,469	1901	do	SO_2, air, mixing and heating of entering gases, and cooling of SO_3 formed.	Apparatus (2 illustrations) consisting of inclosure containing contact body and cooling fluid conduit. The cooling fluid after absorbing the heat of reaction is used to heat the incoming gases.	The gaseous mixture is led through a number of small pipes containing the contact body and thus thoroughly mixed. The heat of the reaction is used to heat the gases entering the tubes.	Economy in fuel, mixing of gases, and control of temperature.
688,470	1901	do	do	Apparatus (2 illustrations) consisting of receptacle for catalyzer and inclosure for cooling medium adjacent to contact chamber, also regulator for controlling temperature of cooling medium.	The incoming gases are heated by the medium which has absorbed the heat from the SO_3 formed. A temperature regulator prevents excessive heating or cooling.	An apparatus for the efficient mixing of the gases and control of their temperature.

TABLE XII.—*Miscellaneous apparatus and processes for the production of SO_3 or H_2SO_4 by the contact process.*

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
700,249	1902	Sébellot, A. M. G.....	SO_2, air, steam, spongy platinum, fibrous asbestos, pumice.	Apparatus (2 illustrations) consisting of vertical casing containing a perforated partition at bottom on which is supported a thick layer of asbestos and pumice. A series of plates containing platinized asbestos.	A mixture of air, SO_2, and steam is admitted to the upper part of chamber containing the contact body. The H_2SO_4 produced proceeds downward through the asbestos and pumice stone and is drawn off at the base.	Efficient oxidation of SO_2 and condensation of H_2SO_4.
875,909	1908	Heinz & Chase.....	SO_2, oxides of nitrogen, catalytic agent.	Process (2 illustrations).....	The gases are mixed with the oxides of nitrogen and conducted successively through a dust flue, a Glover tower, and a Gay-Lussac tower. The residual gases are then filtered and heated and passed over some contact mass.	Combination of chamber and contact process, requiring no elaborate purifying devices.

TABLE XIII.—*Apparatus and processes for utilizing the gases from smelters in the manufacture of sulphuric acid by the chamber process.*

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of process or apparatus.
891,116	1908	Tufts, G. J., and Bramard, E. E.	SO_2 supplied from burning S in addition to that from the furnaces.	Apparatus (1 illustration) comprising an equalizing chamber of relatively large area for rendering the SO_2 from the smelters more nearly uniform in temperature and concentration; also a burner for sulphur.	The percentage of SO_2 in the smelter gases is kept constant by admitting either air or SO_2 into the equalizing chamber.	Apparatus for maintaining SO_2 in furnace gases at constant temperature and in constant quantity.
962,493	1910	Channing & Falding..	.....do.....	Process (2 illustrations).....	The gases from a number of furnaces are led into a common chamber or reservoir and mixed. By admitting air or SO_2 from an auxiliary source a mixture of constant composition is maintained.	Production of H_2SO_4 from smelter gases.
1,046,915	1912	Wedge, U.....	SO_2, water, heat.....	Process (1 illustration).....	The gases from the smelters are led over quiescent bodies of water maintained at a comparatively low temperature. The water is then heated and agitated and the SO_2 driven off.	Absorption of SO_2 from smelter gases and its subsequent utilization.

TABLE XIV.—*Apparatus and processes designed to mix and cool the mixture of gases used in the manufacture of sulphuric acid by the chamber process.*

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
1,303	1839	Hargreaves, J.	SO_2, air blast, steam, oxides of nitrogen.	Process (1 illustration).	The furnace gases are blown by an air blast and steam down through a pipe to the bottom of a tank. The gases then pursue a meandering course up through this tank.	Means of making H_2SO_4.
114,042	1871	Ravenel, St. J.	SO_2, air, nitrate of soda, water, cooling action.	Apparatus (1 illustration) consisting of a cooling tower, a chamber, a smaller compartment, and a final tower.	The gaseous mixture is led through the cooling tower, then into the acid chamber, then into the smaller chamber, and finally the last traces of H_2SO_4 are removed in the last tower.	The method of producing H_2SO_4 with less chamber space.
349,241	1886	Thyss, J. J.	SO_2, air, steam, nitrate of soda.	Apparatus (2 illustrations) consisting of towers fitted at intervals with horizontal perforated lead plates.	The gases are led down through a series of the towers, meeting a steam current. The numerous contacts tend to condense the H_2SO_4 formed.	Combined condensers and acid-forming chambers.
345,140	1886	Hughes, J.	SO_2, air, water spray, oxides of nitrogen.	Process (1 illustration).	The hot furnace gases meet a spray of water, which is thus volatilized. The gaseous mixture is then led through a flue containing water and into a chamber, where H_2SO_4 is formed.	Economy in fuel and efficient method of producing H_2SO_4.
353,222	1886	do.	do.	Apparatus (3 illustrations) consisting of a flue containing water, and chamber for production of H_2SO_4.	do.	Apparatus for carrying out process described in patent No. 345,140.
446,060	1891	Delplace, E. & J.	SO_2, air, oxides of nitrogen, mixing and cooling action, water.	Apparatus (4 illustrations) consisting of annular lead chamber provided with two gas inlets, and containing distributing tubes at intervals to convey the hot gases from the upper to lower part of chamber.	The gases are given a rotary motion, which tends to mix them thoroughly.	More efficient method of producing H_2SO_4 due to better mixing of gases.
485,126	1892	Lunge, G.	Oxides of nitrogen, mixing and cooling action, water, SO_2, air.	Apparatus (8 illustrations) consisting of plates contained in towers so arranged that the gases are continually impinging on a film of cool dilute acid. The acid splashes from one layer of plates to another.	The hot gases are mixed and cooled by ascending the plate column and meeting the dilute H_2SO_4 dripping down the tower.	Apparatus for the cooling and mixing of gases and condensation of H_2SO_4 formed.

TABLE XIV.—*Apparatus and processes designed to mix and cool the mixture of gases used in the manufacture of sulphuric acid by the chamber process*—Continued.

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
546,596	1895	Pratt, N. P.	SO_2, air, oxides of nitrogen, mixing and cooling action, water.	Process and apparatus (6 illustrations). Apparatus consists of an auxiliary cooling tower and means for withdrawing part of the gases from the chambers and reintroducing them into the front of the system.	The gases, after traversing the first chamber, are in part withdrawn and reintroduced into the front of the chamber. The remainder of the gas proceeds to a cooling tower fed with dilute H_2SO_4 and packed with quartz.	Apparatus and process for thoroughly mixing and cooling gases and precipitating H_2SO_4 formed.
598,351	1898	Staub, A.	do.	Apparatus (12 illustrations) consisting of a series of towers packed with especially constructed acid-resisting bodies.	The gases make their way up the towers and are both mixed and cooled by acid-proof packing.	Efficient yield due to character of packing material.
652,687	1900	Pratt, N. P.	do.	Apparatus (2 illustrations) consisting of an acid-resisting fan connected with the flue leading from the Glover tower and with the chambers.	The gases, after traversing the first chamber, are drawn by the fan into the flue leading from the Glover tower and thus mixed with the fresh gases from the furnaces.	Device for accelerating the reactions.
652,689	1900	do.	do.	Apparatus (1 illustration) consisting of acid chamber with flue leading therefrom and connecting with generator of acid-making gases. Two fans are provided for withdrawing gases and reintroducing them into system.	The partially spent gases are reintroduced into the front of the system and discharged against the current caused by an accelerated draft.	Apparatus for thoroughly mixing gases and accelerating reactions.
652,690	1900	do.	do.	Apparatus (3 illustrations) consisting of blower connected with rear of systems and also with Glover tower, and a flue for reintroduction of partially spent gases, which are mixed with the fresh gases from the furnace.	The partially spent gases and the fresh gases from the Glover tower are mixed and blown into the first chamber.	Do.
688,538	1901	Meyer, T.	do.	Apparatus (2 illustrations) consisting of circular chamber with gas inlet near the top of side wall and outlet at center of chamber bottom.	The mixture of gases is admitted at a tangent and pursues a circular spiral course. The spent gases gradually sink toward the center of the chamber's base and are withdrawn.	Apparatus to produce thorough mixture of gases and control temperature.

688,872	1901	.....do.....	.....do.....	Process (2 illustrations) carried out by apparatus described in patent No. 688,538.	.....do.....	Process to produce thorough mixture of gases and control temperature of system.
715,142	1902	Pratt, N. P.....	.....do.....	Process (2 illustrations).....	The reacting gases are cooled by surrounding the flues between the chambers with cooling devices. The uncombined gases are withdrawn from the rear of system and reintroduced with fresh gases into the first chamber.	Efficient method of mixing gases and controlling temperature of these gases.
728,914	1903	Heinz, N. L.....	.....do.....	Apparatus (4 illustrations) consisting of an arrangement of 2 flues for each chamber. The flues enter the chamber from opposite sides at different levels.	The gases are introduced at opposite sides of the chambers at different levels and made to proceed transversely in their course through the chambers.	Apparatus for more efficient mixing of gases.
736,087	1903	Graham, J. G.....	.....do.....	Apparatus (11 illustrations) consisting of longitudinal trough-shaped contact pieces arranged transversely in tower or chamber.	The gases are passed into a chamber or tower packed with the acid-resisting bodies. The hollow sides of the contact pieces are disposed toward the inflowing gases.	Efficient packing material for towers.
765,834	1904	Hegeler & Heinz.....	.....do.....	Apparatus (1 illustration) consisting of fan in main flue between Glover tower and first chamber; another fan in smaller branch flue for reintroducing gases into Glover tower.	The gases after leaving the Glover tower are in part withdrawn and reintroduced into the Glover tower.	Apparatus for obtaining thorough mixture of gases.
767,335	1904	Evers, R.....	.....do.....	Apparatus (15 illustrations) consisting of denitrating tower, means of providing steam and hot air to said tower, a condensing tower, and means for mixing and cooling gases.	The acid is denitrated in first tower by means of steam and hot air. The gases are cooled, mixed, and the H_2SO_4 condensed in the second chamber.	Combined denitrating and condensing plant.
848,631	1907	Cellarius, R.....	.....do.....	Apparatus (2 illustrations) consisting of turret lined with coke and containing a turbine which is operated by steam-blast.	Gases enter tower at bottom and are thoroughly mixed and the acid formed precipitated by the turbine.	Apparatus for mixing of gases and precipitation of H_2SO_4 formed.
852,390	1907	Liline, I. P.....	.....do.....	Apparatus (3 illustrations) consisting of chamber containing series of vertical air-cooled lead pipes extending from top to bottom. Partition walls having numerous transverse openings are disposed between each set of pipes.	The gases in passing through the chamber are cooled by the lead pipes, the nitrosulphuric acid formed is then decomposed, the H_2SO_4 precipitated, and the oxides of nitrogen thus liberated.	Apparatus for more efficient production of H_2SO_4 at less cost.

TABLE XIV.—*Apparatus and processes designed to mix and cool the mixture of gases used in the manufacture of sulphuric acid by the chamber process*—Continued.

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
860,968	1907	Eliel, O. H.	SO_2, air, oxides of nitrogen, mixing and cooling action, water.	Apparatus (3 illustrations) consisting of several chambers connected in series, the chambers having their greatest extension arranged horizontally. The exit and entrance of chambers are at the same end.	The gases enter the first chamber from the Glover tower in the usual way. A jet of steam in the flue accelerates their velocity. The gaseous steam strikes the rear of the chamber and is thrown back toward the front end, where it is discharged.	Efficient mixture of gases by means of apparatus described.
1,022,493	1912	Meigs, C. C.	SO_2, air, oxides of nitrogen, water, compression and expansion of gases, cooling action.	Apparatus (2 illustrations) consisting of chambers containing air-cooled deflector.	The burner gases enter a rectangular lead chamber and striking the concave sides of an air-cooled deflector are thrown against the sides of the chamber and into the currents of gas passing toward rear of chamber.	Apparatus for thoroughly cooling and mixing reacting gases.
1,057,149	1913	Heinz, N. L.	Oxides of nitrogen, water, and air introduced at intervals into system.	Process (2 illustrations)	The gaseous mixture and steam are treated, in passing through the system, with such regulated quantities of air introduced at intervals that a uniform pressure and movement of the gases is maintained and an amount of oxygen supplied equal to that consumed by the reactions.	Process of accelerating and completing reactions in the chambers.
1,104,590	1914	Wedge, U.	Oxides of nitrogen, water, cooling action.	do	The gases are led at high speed through a plurality of lead chambers or towers of relatively small cross-section and presenting 310 feet of cooling surface for each cubic foot of chamber space.	Process for oxidizing SO_2 to H_2SO_4 in ho diluted furnace gases.
1,112,424	1914	McFarlane, E. H.	do	Apparatus (1 illustration) consisting of blower and device responsive to variations in volume of gases and controlling speed of blower.	A forced draft is induced in the gaseous mixture by a blower, the speed of which is automatically controlled by the volume of the gases passing through the system.	Apparatus for accelerating reactions in lead chambers.
1,112,546	1914	Mills, W., and Packard, C.	do	Apparatus (1 illustration) consisting of a chamber shaped like a frustum.	The chamber is so arranged that water flows over the outside walls, thus cooling the reacting gases.	Apparatus for cooling chamber gases.

TABLE XV.—*Apparatus and processes designed primarily to effect economy in the consumption of niter used in the manufacture of sulphuric acid by the chamber process.*

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
292,054	1884	Rigby, J. S.	Solution of $NaNO_3$, H_2SO_4.	Apparatus (1 illustration) consisting of niter pots into which H_2SO_4 and a solution of $NaNO_3$ can be led.	By regulating the supplies of acid and $NaNO_3$ a steady stream of oxides of nitrogen is furnished to the acid system.	Apparatus for providing oxides of nitrogen in proper quantities.
292,078	1884	Walsh, M. A.	None.	Apparatus (3 illustrations) consisting of an improved Glover tower so arranged that the descending acid can not collect in any pockets and act as an absorber for freshly freed oxides of nitrogen.	The tower and packing so arranged that there are no pigeonholes in which cooler acid can collect and absorb the oxides of nitrogen set free by the hot-burner gases.	Improvement in construction of Glover tower and consequent saving of niter.
350,416	1881	Benker, F., and Lasne, H.	N_2O_4, SO_2, air, water.	Process (no illustrations).	The gases after leaving the lead chambers are mixed with SO_2, which reduces the N_2O_4 to N_2O_3, the latter compound being readily absorbed in the Gay-Lussac tower.	Economy in niter consumption.
375,121	1887	Chappell, F. W.	None.	Apparatus (8 illustrations) consisting of a series of cups (lead) supported from framework of towers and attached to lead top by distributing pipes. Inverted glass tumblers placed in these cups on spiders form hydraulic seals.	The pipes which form the communication between the cups and the tower project above the base of the former, thus forming a ring in which the acid collects. The glass tumblers inverted and placed in these rings form the hydraulic seal. The overflow from the rings runs into the tower.	An apparatus for the uniform distribution of acid to the Glover and Gay-Lussac towers.
378,289	1888	do.	None.	Apparatus (8 illustrations) consisting of a reactionary wheel which distributes acid against a ring which in turn distributes it to the various pipes. (Used in connection with apparatus described in patent No. 275,121.)	The reactionary wheel in revolving uniformly distributes the acid to the various pipes feeding the tower.	Apparatus for uniformly distributing acid to Glover and Gay-Lussac towers, and consequent saving of niter.

388,406	1888	Frazier, S.	None	Apparatus (1 illustration) consisting of a reactionary wheel provided with emergency distributors for any excess of acid which may be supplied to the apparatus.	The reactionary wheel in revolving uniformly distributes the acid to the pipes feeding the tower. By means of emergency distributors the supply of acid can be greatly increased.	Do.
659,236	1900	Johnson, A. C.	Oxygen-hydrogen mixture in proportions to form water, SO_2, air.	Process (3 illustrations)	The gaseous mixture is passed into a chamber in the usual way. Oxygen and hydrogen are also introduced into this same chamber and continually exploded by means of an electric spark.	Method of forming and precipitating H_2SO_4 without use of oxides of nitrogen.
725,427	1903	Eliel, O. H.	None	Apparatus (4 illustrations) consisting of new form of Glover tower having three separately packed sections	The gases from the furnace or burners are drawn down through the tower in three streams along with the sulphuric acid to be denitrated and concentrated.	Improvement in construction of Glover tower, resulting in saving in niter consumption.
755,247	1904	Salom, P. G.	Electric current, SO_2, air	Process (1 illustration)	The SO_2 and air are passed through a series of independent but communicating receptacles each of which constitutes an electrolytic cell. Oxidation takes place and the H_2SO_4 formed is then drawn off.	Process for making H_2SO_4 without the use of oxides of nitrogen.
850,820	1907	Derrig, P. J.	$NaNO_3$ solution, H_2SO_4, SO_2, air.	Apparatus (2 illustrations) consisting of lead shell packed with quartz or coke down which a solution of $NaNO_3$, and H_2SO_4 flows.	Part of the gases from the pyrites burners is made to pass through this coke column, thereby being nitrated.	Apparatus for nitrifying SO_2 used in chamber process.
898,390	1908	Pauling, H.	Dilute HNO_3, nitrosulphuric acid.	Process (1 illustration)	The anode compartment of a suitable electrolyzer is filled with nitrosulphuric acid and the kathode compartment with dilute HNO_3. On electrolyzing the liquid the oxides of nitrogen liberated at the kathode are led into and absorbed in the anode chamber.	Process for regeneration and concentration of dilute acid containing oxides of nitrogen.
900,688	1908	Bender, O.	SO_2, O, and N_2, carbonaceous fuel, oxyhydrogen flame.	Process (2 illustrations)	The gaseous mixture (no oxides of nitrogen) is led through a carbonaceous fire. Partly dissociated steam is then introduced into the mixture and the free H_2 and O in combining heat the gases still higher. The gases are then cooled by steam.	Production of H_2SO_4 without niter in small chamber space.
904,147	1908	Petersen, H.	Cold nitrous vitriol (55° B.), SO_2, air, water.	do	The gases after leaving the last chamber are brought into contact with cold nitrous vitriol (55° B.) before proceeding to the Gay-Lussac tower.	Process for preventing escape of SO_2 and oxides of nitrogen.

TABLE XV.—*Apparatus and processes designed primarily to effect economy in the consumption of niter used in the manufacture of sulphuric acid by the chamber process*—Continued.

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
908,696	1909	Petersen, H.....	Two supplies of nitrous vitriol of different strengths and two supplies of Glover acid of different strengths, SO_2, air, water.	Process (5 illustrations).....	The gaseous mixture is passed into contact with two supplies of nitrous vitriol, then through the chambers in the usual way, and finally in contact with two supplies of Glover acid of different strengths.	Efficient method of oxidizing SO_2 and recovering oxides of nitrogen.
1,068,021	1913	Tarand, A., and Truchot, P.	Water solution of Na_2CO_3, SO_2, air, water.	Process (1 illustration).....	The gases after leaving the Gay-Lussac tower are led up through a tower packed with acid-resisting material down which water trickles. The gases are then led successively through two towers into which a solution of Na_2CO_3 is sprayed. The $NaNO_3$ resulting is then sprayed into the chambers.	Process for recovery of last traces of oxides of nitrogen and sulphuric acid.

TABLE XVI.—*Apparatus and processes designed primarily to reduce the amount of chamber space required in the manufacture of sulphuric acid by the chamber process.*

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of process or apparatus.
86,881	1869	Tait, A. H	Liquid SO_2 and NO_2 free from nitrogen, steam.	Process (4 illustrations)	The SO_2 from the pyrites burners is liquefied and thus freed from N; it is then introduced together with NO_2 under pressure into a chamber into which air and steam are also forced. The NO_2 is oxidized to NO_3, which is liquefied, and mingling with the liquid SO_2, oxidizes the latter.	Efficient production of H_2SO_4 in small amount of chamber space.
143,202	1873	Thomson, E., and Green, W.	SO_2, air, nitric acid, cooling.	Apparatus (5 illustrations) consisting of two towers, one for the reactions and the other for absorbing the oxides of nitrogen.	The burner gases are admitted at two points in the reaction tower which is fed with HNO_3. The condensed acid is drawn off and the oxides of nitrogen absorbed from the residual gases by passing through an absorption tower fed with water.	Production of purer and more concentrated H_2SO_4 with less chamber space.
535,882	1895	Barbier, E. J	HNO_3, SO_2, air, heat	Apparatus (5 illustrations) consisting of three reaction towers containing piles of perforated pots, and one Gay-Lussac tower.	The gases are led from the furnace through three towers filled with perforated pots and fed with HNO_3. The base of each tower consists of a perforated plate under which there is an acid pan heated by a separate fire.	Efficient apparatus for producing H_2SO_4 without use of large chambers.
652,688	1900	Pratt, N. P	SO_2, air, oxides of nitrogen, cooling and mixing.	Apparatus (1 illustration) consisting of two series of towers (in lieu of chambers) packed with some suitable acid-resisting bodies. A flue connects the first of the second series with the last of the first series. A blast mechanism is contained in the flue.	The gaseous mixture is drawn through the first series of packed towers or converters. Part of the gas then goes to the Gay-Lussac tower and part is drawn through a second series of converters by means of a flue connecting the two series.	Substitute for lead chambers.
729,643	1903	Neumann, Max	SO_2, air, oxides of nitrogen, water heating and cooling.	Process (5 illustrations)	The furnace gases are drawn through a plurality of Glover towers and through alternating heating and cooling zones intermediate of said towers.	Efficient production of H_2SO_4 without employing lead chambers.
765,520	1904	Stinville, A. L	SO_2, air, oxides of nitrogen, H_2SO_4 (3° to 5° B. below that produced on chamber walls), cooling.	Process (1 illustration)	Cool dilute H_2SO_4 in the bottoms of a plurality of chambers serves to cool and condense H_2SO_4 formed, and to furnish part of water required for system.	Economy in chamber space and cost of construction.

TABLE XVI.—*Apparatus and processes designed primarily to reduce the amount of chamber space required in the manufacture of sulphuric acid by the chamber process*—Continued.

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
932,771	1909	Falding, F. J.	SO_2, air, oxides of nitrogen, water.	Apparatus (6 illustrations) consisting of the usual chamber plant, except that the chambers are one and one-half times higher than their horizontal dimensions and are not connected in series.	The mixture of gases is led from the Glover tower into the lead chambers near the top. A hot zone of the reacting gases is thus formed at the top of the chambers. The spent gases gradually settle to the bottom and are withdrawn.	Efficient production of H_2SO_4 due to the segregation of the reacting gases from the spent gases.
1,012,421	1911	Opl, C.	Nitrosulphuric acid. SO_2, water, compressed air.	Process (2 illustrations).	Part of the burner gases, together with compressed air, is made to lift the nitrosulphuric acid from the acid eggs, said acid is then sprayed from the tops of a number of reaction towers through which the bulk of the burner gases is led.	Production of H_2SO_4 without use of ordinary lead chambers, thus economizing in chamber space.
1,048,953	1912	Fulda, W.	SO_2, air, HNO_3, water.	Process (1 illustration).	The burner gases are treated with HNO_3. The reduced HNO_3 is then regenerated by passing down a plurality of oxidizing towers.	Production of H_2SO_4 without use of lead chambers (ordinary) and without loss of HNO_3.

TABLE XVII.—*Apparatus and processes designed primarily for the concentration of the sulphuric acid manufactured by chamber process.*

Patent No.	Year.	Patentee.	Reagent used.	Apparatus employed.	Treatment.	Object of process or apparatus.
541,041	1895	Falding, F. J.	Heat of burner gases	Process and apparatus (6 illustrations) consisting of concentrating tower.	The acid from the Glover tower is allowed to trickle down an absorption tower filled with some suitable packing material.	Production of more concentrated H_2SO_4.
699,011	1902	Quinan, W. R.	do	Apparatus (6 illustrations) consisting of a concentrating flue arranged in a series of steps.	The acid from the Glover tower flows down the flue in a thin stream, being exposed to the action of the burner gases, the heat of which concentrates the acid.	Production of H_2SO_4 of 66° B.
752,677	1904	Heinz, N., and Hegeler, H.	Oxides of nitrogen, SO_3, heat of burner gases.	Process (2 illustrations)	The gases are withdrawn from any part of the system and mixed with the gases entering the Glover tower.	Production of more concentrated H_2SO_4 due to absorption of SO_3 from gases.
768,108	1904	Zanner, Adolf	Heat of burner gases	Apparatus (3 illustrations) consisting of an enlarged flue (containing vessels of H_2SO_4) between the burners and the Glover tower.	The heat of the burner gases is utilized to concentrate H_2SO_4. The latter exerts a cooling action on the gases and also furnishes water to the system.	Production of more concentrated H_2SO_4.
963,174	1910	Proelss, O.	do	Process (3 illustrations)	The burner gases are conducted in two independent streams through a denitrating apparatus and a concentrator. The two streams are then brought together and passed through a cooler in contact with dilute H_2SO_4 containing oxides of nitrogen. The gases are finally admitted to the chambers.	Production of concentrated H_2SO_4.
963,175	1910	do	do	Process (1 illustration)	The burner gases are conducted through a concentrating tower supplied with ordinary H_2SO_4. The concentration of the acid is carried to the point where the impurities are deposited.	Production of more concentrated and purer H_2SO_4.
989,537	1911	do	do	Apparatus (2 illustrations) consisting of a plurality of towers for concentrating H_2SO_4, and means for supplying acid to said chambers, also cooling apparatus.	Apparatus for carrying out process described in patent No. 963,175.	Apparatus for production and concentration of H_2SO_4.

TABLE XVIII.—*Miscellaneous apparatus and processes for the manufacture of sulphuric acid by the chamber process.*

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
130,432	1872	Jackson, D	SO_2 from S, HNO_3, air, water.	Apparatus and process (4 illustrations) consisting of a special furnace for burning S and chambers containing screens to filter out fine S.	The mixture of gases and finely divided S are led through a chamber containing screens. The gases then go to the denitrating chamber, where their heat drives off the oxides of nitrogen from H_2SO_4 already formed. The gases then go to the conversion chamber.	Efficient production of H_2SO_4 from S by apparatus described.
150,095	1874	Sprengel, H	Water or acid in form of spray.	Process (no illustrations)	Water or acid is sprayed into the reaction chambers instead of steam.	More efficient method of precipitating the sulphuric acid formed.
230,501	1880	Sébilot, A. G	Ore treated with sulphuric acid, heat, air, steam.	Apparatus (2 illustrations) consisting of a special furnace.	The ore is treated with H_2SO_4 and heated in a furnace, resulting in the formation of sulphates and evolution of SO_2 and H_2SO_4. The SO_2 is oxidized by mixing with air and steam.	Recovery of H_2SO_4 from treatment of ores.
233,680	1880	Labois, E. & L	Heat	Process and apparatus (3 illustrations) for the production of CS_2 and H_2SO_4.	The pyrites are first heated and 15 per cent of their sulphur content driven off; the remaining material is then dumped into a grate and the SO_2 burned out.	A method and process for the production of CS_2 and H_2SO_4 from pyrites.
268,793	1882	Haworth, E	Absorption by and subsequent drawing off of gases from water.	Process (2 illustrations)	The mixture of gases is allowed to bubble through water. The solution of SO_2 is then heated by a steam coil and thus driven off. The water is returned to the first tank and used again.	Purification of SO_2.
270,763	1883	Dotterer, T. D	SO_2, air, steam, water	Apparatus (1 illustration) consisting of a tower connected with a pipe which leads into a tank; the tank is connected with a fume chamber which opens under a pile of coke.	The gases after leaving the lead chamber are drawn up through a long pipe (into which a jet of steam is played) into a tank sprayed with water. The vapors escaping condensation are then led down into a fume chamber and up through a hollow coke pile sprayed with water.	Apparatus for the absorption and condensation of injurious fumes.
308,289	1884	Terrell, T	S and $FeSO_4$, heat	Process (no illustrations)	A mixture of S and $FeSO_4$ is first heated and then burned, resulting in the formation of SO_2, SO_3, and Fe_2O_3. The SO_2 is then converted into H_2SO_4 in the usual way.	Efficient method of making Fe_2O_3 from $FeSO_4$ and H_2SO_4.

TABLE XV III.—*Miscellaneous apparatus and processes for the manufacture of sulphuric acid by the chamber process*—Continued.

Patent No.	Year.	Patentee.	Reagents used.	Apparatus employed.	Treatment.	Object of apparatus or process.
325,262	1885	McNab, J.	Oxides of nitrogen supplied from auxiliary tower.	Process (2 illustrations)	Nitrous vitriol is fed into a tower down which it trickles, meeting a current of steam which denitrifies it. The oxides of nitrogen thus freed are led into the last chamber or to any other.	Method of restoring sick or pale chambers quickly.
325,263	1885	do.	do.	Apparatus (2 illustrations) consisting of auxiliary tower for carrying out process described in patent No. 325,262.	do.	Apparatus for restoring sick or pale chambers quickly.
357,107	1887	Sprengel, H. J. P.	Waste steam	Process (1 illustration)	Waste steam from the exhaust pipe is run into the lead chamber to supply the water necessary in the formation of H_2SO_4 by the chamber process.	Utilization of waste steam and a consequent saving of fuel.
477,755	1892	Hanisch, G. E.	Some salt which does not react with H_2SO_4, such as Na_2SO_4 or $CaSO_4$.	Process (no illustrations)	The H_2SO_4 is mixed with the salt in the quantity necessary to have the latter take up water from the former sufficient to produce a crystallized mass.	Solidification of acid to facilitate shipment of same.
694,024	1902	O'Brien, A. P.	Heat	Apparatus (3 illustrations) consisting of niter oven so arranged in a casing that the latter also serves as a dust chamber.	The niter ovens are heated by means of the furnace gases. Means are provided for drawing off the niter cake.	Niter oven which serves as dust chamber, and which can be readily cleaned.
798,524	1905	McNab, J.	None	Apparatus (14 illustrations) consisting of improved roasting kiln for pyrites.	Kiln is constructed in such a way as to avoid danger of collapsing.	Improved furnace for burning pyrites.
825,057	1906	Johnson, W. McA.	Sulphuric acid in electrolytic cell, SO_2.	Process (3 illustrations)	An electric current is passed through dilute sulphuric acid into which SO_2 is led.	Method of obtaining H_2SO_4 without employing oxides of nitrogen or other catalytic agent.
873,070	1907	Nibelius, A. W.	A volatile solvent for H_2SO_4.	Process (1 illustration)	Niter cake is ground and treated with a volatile solvent for H_2SO_4. After filtering, the volatile substance is distilled off.	Process of obtaining H_2SO_4 from niter cake.
908,400	1908	Eggleston, J. E.	H_2S and means for burning it and oxidizing resulting SO_2.	Apparatus (2 illustrations) consisting of burner for H_2S in connection with a series of oil stills and means (chamber and Glover tower) to oxidize SO_2.	Oil containing sulphur is passed through a series of stills. The H_2S evolved is oxidized to SO_2 and the SO_2 then treated by the chamber process.	Apparatus for separating and oxidizing sulphur contained in oil.

928,844	1909	De Brailles, G. C.....	H_2SO_4, SO_2, under pressure, granules of coke and antimonated lead packed in the inner compartment of a two-chamber cell, electric current.	Process (1 illustration).....	A mixture of H_2SO_4 and SO_2 under pressure and at 45° B. is run into both chambers. An electric current is passed through the cell, which decomposes the water contained in the acid mixture. The hydrogen from the cathode reduces the SO_2, and the oxygen at the anode oxidizes the SO_2 to H_2SO_4.	Production, concentration, and purification of H_2SO_4.
909,578	1909	Gaillard, A.....	Water in form of spray and means to introduce it into system.	Apparatus (1 illustration) consisting of a lead chamber having circular holes cut in its top into which are sealed lead cones. Jet nozzles passing inwardly through said cones, and receptacles below the cones to catch condensed water.	The height of the water sprays from the bottom of the chamber is thus increased, and the condensed water being collected in the receptacles below the cones is led out of the chamber without diluting the H_2SO_4.	Apparatus for introducing water into the chamber.
981,103	1911	Moritz, R.....	None.....	Apparatus (8 illustrations) consisting of framework and means to support acid chambers thereto.	The framework is so constructed that it does not come into contact with the chamber's sides.	Improved construction of chambers.
1,018,040	1912	Eggleston, J. E.....	Heat, steam, H_2S, and means to oxidize latter.	Process (2 illustrations).....	Petroleum containing sulphur compounds is first heated to 300° to 500° F. in a series of stills and then exposed to action of steam. H_2S evolved is burned and treated by chamber process.	Process for utilizing sulphur in petroleum.
1,018,374	1912	Robinson, C. I.....	Heat, H_2S, and means to oxidize latter.	Process (1 illustration).....	Petroleum containing sulphur is heated to about 500° F. The volatile products are passed through a condenser. The H_2S evolved is collected in a gasometer and subsequently burned to SO_2 and oxidized in chambers.	Process for utilizing sulphur in crude oil and purifying latter.
1,106,999	1914	Wedge, W.....	Acetate of an alkali or some other metal in acid solution, iodine solution.	Process (no illustrations)....	The gases are tested before and after entering and leaving the chambers for their content of SO_2 and the proportion of this gas to the other ingredients thus regulated.	Process for determining the amount of SO_2 in chamber gases in order to regulate same.

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